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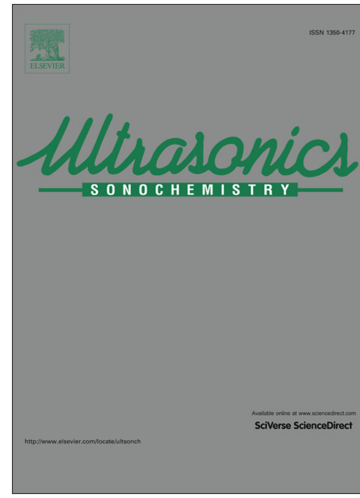

Review

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Simulation of the spatial distribution of the acoustic pressure in sonochemical reactors with numerical methods: A review

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Abstract

Numerical methods for the calculation of the acoustic field inside sonoreactors have rapidly emerged in the last 15 years. This paper summarizes some of the most important works on this topic presented in the past, along with the diverse numerical works that have been published since then, reviewing the state of the art from a qualitative point of view. In this sense, we illustrate and discuss some of the models recently developed by the scientific community to deal with some of the complex events that take place in a sonochemical reactor such as the vibration of the reactor walls and the nonlinear phenomena inherent to the presence of ultrasonic cavitation. In addition, we point out some of the upcoming challenges that must be addressed in order to develop a reliable tool for the proper designing of efficient sonoreactors and the scale-up of sonochemical processes.

Keywords: Acoustic pressure, Numerical Simulations, Sonochemical reactor, Sonochemistry, Ultrasound

1. Introduction

Sonochemistry [1] is the area of high-energy chemistry which studies chemical reactions and processes involving acoustic cavitation formed by the application of an ultrasonic field in a frequency range which commonly varies between 20 kHz and 10 MHz. It allows chemists to increase the conversion, improve the yield, initiate and change the reaction pathways in all sorts of biological, chemical or electrochemical processes [2], becoming a prominently used technique in a wide variety of research areas, including: (i) material science [3] (ii) synthetic chemistry [4, 5], (iii) water remediation [6, 7], (iv) biotechnological applications [8], (v) electrochemical processes [9], (vi) food technology [10], and (vii) spent nuclear fuel reprocessing [11], among others. The versatility of the ultrasonic permits its combination with other technologies such as photocatalysis [12] or microwaves [13], proving the enormous potential of Sonochemistry.

Despite this extensive research at laboratory scale, a limited number of applications have been industrially scaled-up due to two main reasons: (i) the lack of expertise in diverse areas such as ultrasonics or sonochemical engineering, and (ii) the lack of proper reactor designing strategies. Related to this, Sutkar and Gogate have stated that understanding the cavitational activity and its distribution would yield efficiently designed sonochemical reactors and systems [14], and for this purpose, theoretical analysis of the cavitational activity distribution with proper experimental validation could be used for the optimization of sonochemical processes taking into account building materials, geometry of the reactor and working frequency of the sonochemical system. A correct understanding of the acoustic field structure inside a sonochemical reactor is therefore needed to proceed with its optimization and scale-up in order to design efficient large scale reactors [15].

The numerical simulation of the spatial distribution of the acoustic pressure inside sonochemical reactors has widely emerged in the last 15 years to shed new light on this issue, and quite a few groups around the world have tried to model the acoustic field inside sonoreactors with the aim of predicting the cavitation events within the reactor. To our knowledge, the most recent review on this topic found in the literature was published more than 10 years ago [16], and no exhaustive literature revisions are usually found in most of the papers that deal with the simulation of the acoustic field in a sonoreactor. Therefore, the goal of the present paper is to introduce numerical methods for the development of sonochemical reactors to a wider audience of scientists by summarizing the continuous development of numerical methods employed by the scientific community from the late 1990's until now. In this paper, basic methodologies and results from many works are briefly commented, pointing out the strong and weak points in the most representative cases from a qualitative point of view. And new trends and future challenges on the problem are also discussed.

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2. Numerical simulations of the acoustic field inside sonochemical reactors using linear-based models

2.1. Basic linear-based models

The vast majority of the works dealing with the simulation of the acoustic field inside a sonochemical reactor rely on the resolution of the equations that describe the linear propagation of sound in a liquid. Such equations, which are derived from the linearization of the Euler equations [17], yield the well-known Helmholtz equation for the linear propagation of sound waves:

$$\nabla^2 P + k^2 P = 0 \tag{1}$$

being P the acoustic pressure and $k = (\omega / c_l)$ the wave number, where ω is the angular frequency and c_l is the sound speed of the liquid. The Helmholtz equation can be easily solved by setting the next boundary conditions:

- $P = 0$ for 'infinitely soft' boundaries.
- $\nabla P \cdot \mathbf{n} = 0$ for 'infinitely hard' boundaries, being $\mathbf{n}$ the normal vector pointing outward the liquid.
- $P = P_0$ at the emitter surface of the ultrasonic transducer, being P_0 the amplitude of the wave.

The simplicity of the Helmholtz equation and the boundary conditions defined above facilitate the task of calculating the linear propagation of sound in a liquid by numerical methods. In the last years, the wider availability of commercial FEM (Finite Element Methods) software packages presenting acoustic modules based on the Helmholtz equation have allowed an increasing number of different research groups to employ numerical simulations as a powerful tool to design and characterize sonochemical systems.

Among the different commercial software codes available, COMSOL Multiphysics, formerly known as FEMLAB, has probably been the most employed in recent years, not only because the Helmoltz equation and the different boundary conditions are implemented, but also because it does not require deep knowledge on either advanced acoustics modelling or numerical methods. With this software, Sáez et al. [18] tried to characterize a 20 kHz sonochemical reactor by considering the vessel boundaries as infinitely rigid walls and putting special efforts on the discretization of the domain following the rules by Ihlenburg, Babûska and co-workers [19, 20]. In their work, the simulations were compared with aluminum foil experiments, observing in both cases a main active zone just along the axial direction located at the emitter center. Their numerical results also indicated a gradual decrease in the ultrasonic field activity with the distance from the emitter, which was roughly confirmed by aluminum foil experiments. Klíma et al. [21] faced the optimization of a 20 kHz sonochemical reactor based on the three-dimensional simulation of the acoustic pressure with the same software. Despite the limitations of the linear Helmholtz equation, which does not take into account non-linear wave propagation and generation of transversal elastic waves, and the consideration of the reactor walls as infinitely hard (which may not be realistic enough), the authors demonstrated that a proper design of the reactor geometry could yield an increase in the acoustic intensity due to multiple reflections rather than the fast decrease in intensity commonly observed when increasing distance from the horn tip (Fig. 1). Bargoshadi and Najafiaghdam [22] tried to optimize an ultrasonics dispersion system using multiple transducers, observing that the optimum distance between two transducers would yield a more uniform distribution of pressure antinodes, and Shao et al. employed basic numerical simulations to estimate the agglomeration position of oxidation inclusions and the ultrasonic field propagation in a ultrasonic cell employed in the purification of magnesium alloy melt [23, 24], and the combination of an ultrasonic field with an electromagnetic field for casting AZ80 Mg alloy billets [25]. Kim et al. [26] tried to simulate the temperature and pressure profiles of various solvents at different levels of ultrasonic power by combining COMSOL's acoustic and heat transfer modules. More recently, Thiemann et al. [27] simulated the sound field in a 230 kHz system, observing similar trends in both experimental and numerical works. These simulations, which accounted for partially reflecting boundaries, no only showed the standing wave planes away from the transducer, but also the more complex near field. They also observed that the acoustic pressure amplitude in pressure nodes did not reach zero in the observed standing wave (which indicates the presence of partially absorbing boundaries), being this issue considered by the authors as an hypothetical reason for the cavitation structure experimentally observed.

COMSOL Multiphysics has also been used by different authors in order to solve the Helmholtz equation accounting for damping of the ultrasonic effect by introducing the complex density ρ_c and the complex sound speed c_c:

$$\rho_c = \frac{Z_c k_c}{\omega} \tag{2}$$

$$c_c = \frac{\omega}{k_c} \tag{3}$$

where k_c is the complex wave number and Z_c is the complex impedance. Different expressions can be found in the literature for Z_c and k_c. For example, we can find that

$$k_c = \frac{\omega}{c_l} \frac{1}{\sqrt{1 + \left(i\omega\mu_l / \rho_l c_l^2\right)}} \tag{4}$$

$$Z_c = \rho_l c_l \frac{1}{\sqrt{1 + \left(i\omega\mu_l / \rho_l c_l^2\right)}} \tag{5}$$

where μ_l and ρ_l are the viscosity and density of the liquid, respectively. These expressions were used by Sutkar et al. in order to simulate the acoustic field in diverse low [28] and high frequency [29] sonoreactors. A similar approach was recently followed by Xu et al. [30], although in this latter case $k_c = (\omega/c_l) - i\alpha$ and $Z_c = \rho_l c_l$, where α is an attenuation coefficient analogous to used by other authors later mentioned in this paper [44, 47, 34, 62, 63]. In this recent work, the authors simulated a high frequency sonoreactor accounting for the fountain effect commonly observed on the liquid surface in such type of sonochemical systems by roughly adjusting the geometry simulated. They also estimated the liquid flow within the reactor.

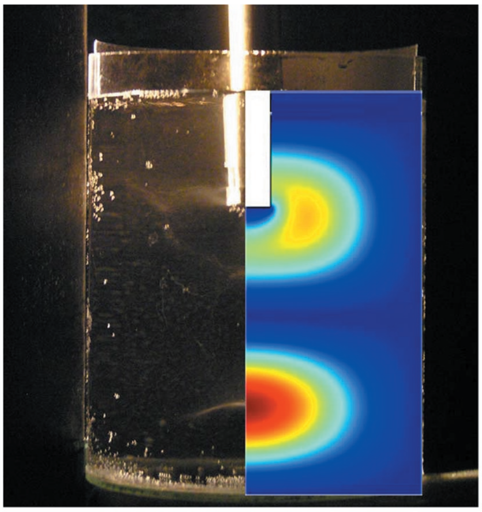

Figure 1: Photograph of cavitating bubbles in a 20 kHz optimized cell (left) and simulated intensity distribution for the same geometry (right). Reprinted from [21], with permission from Elsevier.

Besides COMSOL Multiphysics, many other FEM codes have also been used in recent works dealing with the simulation of the acoustic field inside a sonoreactor. The ATILA code, which has been employed for many years in the analysis of 2D and 3D geometries in different engineering fields involving linear acoustics, vibrational mechanics and piezoelectricity (being widely used for underwater acoustics applications [31]), has been used by Perincek et al. [32] to evaluate the spacing and alignment of the piezoelectric transducers in an ultrasonic bath in order to design efficient industrial bath for textile treatment purposes (no details of the resolutions of the linear acoustics model were included in the paper). The PZflex code has also been used in different cases but to a lower extent, as in the work from Mutasa et al. [33] focused in the estimation of the pressure field in cylindrical reactors of different sizes. Dahlem et al. [34], in their attempt to simulate the acoustic streaming above the cavitation threshold, coupled the Sysnoise and Fluent codes to model both the ultrasonic pressure field and the velocity field. The authors calculated the driving force used as the source term in their Computer Fluid Dynamics (CFD) code from the acoustic pressure and sound velocity fields estimated with the Sysnoise software. In this case, not much information on the linear model used was given (the resolution of the hydrodynamics side of the problem was based on the theory of fast acoustic streaming [35]). And Laborde et al. [36] dealt with the prediction of the acoustic cavitation field in low and high frequency sonoreactors employing two different mathematical approaches: (i) the resolution of Euler equations with the Eole code to solve the equations that describe the linear propagation of sound waves throughout a still and incompressible liquid, and (ii) the resolution of Navier-Stokes equations coupled with the energy equation, which were solved along with the equation of state of the fluid by CFD code Aquilon in order to describe the sound field in a more complex compressible flow where only thermal exchanges by conduction are considered. For low frequency simulations compared with aluminum foil experiments, Eole simulations provided unrealistic pressure fields while Aquilon seemed to give better results. However, for high frequency/low power simulations the Eole method gave pressure fields which were in agreement to some extent with sonochemiluminescence experiments with luminol, as nonlinear acoustic terms could be neglected and cavitation bubbles were small and homogeneously spread through the reactor (low attenuation in the acoustic field inside high frequency-low power sonochemical reactors is usually observed in experiments). Nevertheless, they commented that acoustic nonlinearities would appear along with an increase in convection due to the radiation pressure at higher power, where the Euler-based model would no longer be suitable.

2.2. Linear-based models accounting for the presence of bubbles

The resolution of the linear equations on their own, although may actually lead to numerical results that agree to some extent with certain experimental observations due to the simplicity of the geometry simulated (Fig. 1), is far from describing the complex nature of the acoustic field in real sonochemical reactors and it does not take into account the attenuation of the propagation of sound due to the presence of the cavitating bubbles (although a few of the works previously mentioned did account for wave attenuation to some extent by considering complex parameters [28, 29] and including an absorption coefficient of the acoustic wave [30, 34]). It is therefore necessary to account for the effects that the presence of the bubbles have on the propagation of sound inside a sonoreactor. Wave propagation in a bubbly liquid has been theoretically studied for many years, being much of the theoretical work on the field developed around a set of nonlinear equations proposed by van Wijngaarden [37, 38, 39]. Such set of equations were derived by Caflisch et al. [40] by adapting Foldy's method [41, 42]. Commander and Prosperetti [43] developed a linear model of the so-called Caflisch equations for both mono and polydisperse bubble populations. The linear nature of the latter model is of great interest in the simulation of sonochemical reactors, as it allows the straightforward incorporation of wave attenuation into the Helmholtz equation by employing a complex wave number k_m instead of wave number k in equation (1), being k_m defined as:

$$k_m^2 = \frac{\omega^2}{c_l^2}\left(1 + \frac{4\pi c_l^2 n_b a_0}{\omega_0^2 - \omega^2 + 2ib\omega}\right) \tag{6}$$

where n_b is the number of bubbles per unit volume, a_0 is the equilibrium radius of bubbles in monodisperse distribution, ω_0 is the resonance frequency of the bubbles, i is the imaginary unit and b is the damping factor, which is defined as:

$$b = \frac{2\mu_l}{\rho_l a_0^2} + \frac{P_b}{2\rho_l \omega a_0^2} \Im \Phi + \frac{\omega^2 a_0}{2c_l} \quad (7)$$

being μ_l the viscosity of the liquid, P_b the undisturbed pressure at the bubble location and $\Im$ the imaginary part of a complex number. The complex dimensionless parameter Φ is defined as:

$$\Phi = \frac{3\gamma}{1 - 3(\gamma - 1) i\chi \left[(i/\chi)^{1/2} \coth (i/\chi)^{1/2} - 1 \right]} \quad (8)$$

where γ is the specific heat ratio of the gas inside the bubbles and $\chi = D/\omega a_0^2$, being D the thermal diffusivity of the gas. The real part of the complex dimensionless parameter Φ is also used to calculate ω_0 as follows:

$$\omega_0^2 = \frac{P_b}{2\rho_l a_0^2} \left(\Re \Phi - \frac{2\sigma_l}{a_0 P_b} \right) \quad (9)$$

where σ_l is the surface tension of the liquid. These equations allow to define the volume fraction of bubbles in the reactor β as follows:

$$\beta = \frac{4\pi}{3} n_b a_0^3 \quad (10)$$

The linearization of the Caflisch equations proposed by Commander and Prosperetti [43] result in a powerful tool in the simulation of the acoustic field inside sonochemical reactors, as it enables the simulation of the acoustic field in a sonochemical reactor accounting for the effect of the presence of the bubbles to some extent. As an example, we can comment the work by Dähnke and Keil. Initially, they developed a model to calculate the three-dimensional distribution of the linear acoustic pressure in liquid media with an homogeneous and inhomogeneous distribution of bubbles [44] considering a combination of the Helmholtz integral [45] and the Kirchhoff integral equations [46]. In this model, they introduced Commander and Prosperetti's linearization of the Caflisch equations in order to calculate the attenuation coefficient and phase velocities by taking into account the bubble volume fraction β. Therefore, they could relate in their model the acoustic pressure in the reactor to the presence of the cavitating bubbles. In this work, the time-independent acoustic field was modelled stepwise in the beam direction, with the bubble density distribution being calculated every step considering the phase velocity and the attenuation coefficient, observing that inhomogeneous density distributions of cavitation bubbles remarkably affected the propagation of the ultrasonic waves and the spatial distribution of the acoustic pressure. They observed that damping effects were dominating and cavitation was restricted to the vicinity of the beam source when the initial pressure amplitudes set at the emitter boundary reached certain values. Such model was further developed in a second work [47], where the authors were able to take into account the density distribution of bubbles depending on the spatial distribution of the acoustic pressure amplitude in order to solve the time-dependent wave equation in a more realistic approach by assuming that bubbles form and grow near pressure antinodes, which are the areas where maximums of acoustic pressure are reached. Further works by the same authors using the finite difference approach included the simulation of different reactor configurations [48, 49], comparing the numerical results with experimental data obtained by Soudagar and Samant in a previous work [50]. Although they found similar trends in the spatial distribution of the acoustic field (such as the 'shape' or structure of the acoustic field), quantitative agreement was not observed. More recent works have also implemented Commander and Prosperetti's linearization of the Caflisch into linear-based acoustics following a similar approach [51, 52], being the latter of them of certain interest for the audience as the authors, Jordens et al., tried to combine acoustics, hydrodynamics and reaction kinetics in the simulation of ultrasonic reactors with confined channels. Despite some serious limitations of the simulations already pointed by the authors (they considered a laminar flow profile with incompressible flow assuming no interaction between the ultrasonic field and the liquid flow), their simulations may be used to suggest the modification of different process parameters such as ultrasonic power and frequency, geometry, etc. in order optimize the real reactor.

The Caflisch equations themselves have also been incorporated into different models in order to account for the effect of the presence of the bubbles on the distribution of acoustic pressure in sonochemical reactors. Servant et al., in a further development of their previous model [36], simulated cavitation bubble dynamics in a high frequency sonoreactor [53]. Once again, they used the Aquilon code to solve Euler and Navier-Stokes equations, with a modification of the code to implement the Caflisch equations. The employed model not only determined the bubbles' emergence sites within the reactor, which resulted in the migration of transient cavitation bubbles towards pressure nodes, but also the motion of the fluid and the time-averaged velocity field of the bubbles. This model was later improved by considering bubble density as a time-dependent variable through the bubble volume fraction, which was assumed to be linearly dependent on the acoustic pressure amplitude [54]. Even though the authors separately computed the acoustic wave propagation and the migration of the bubble clouds (under the rough assumption of no interaction between the bubbles and the sound field, which indeed is not realistic [55]), they obtained qualitative results similar to those experimentally observed with aluminum foil tests as shown in Fig. 2. The same authors also extended the model to calculate the interaction between the acoustic field and the bubble clouds in dual frequency reactors [56] by using the CAMUS code.

Even though the implementation of the Caflisch equations in their linear version in linear-based acoustics constitutes a more rigorous approach to the problem that the mere use of basic linear equations such as the Helmholtz equation, it may not be enough to realistically simulate how cavitation phenomena really affect the acoustic field inside a sonochemical reac-

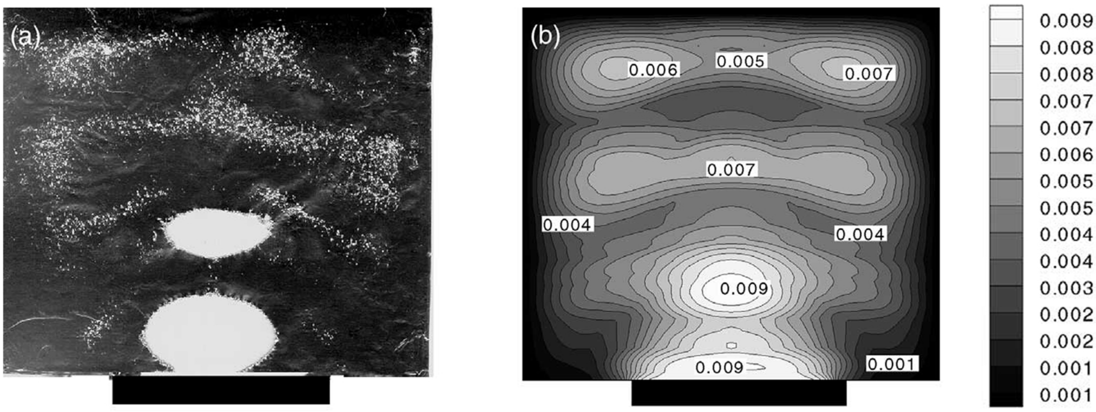

Figure 2: (a) Longitudinal view of aluminum foil erosion (exposure time = 90 s), and (b) computed volume fraction of fragmentary transient cavitation bubbles in a 28.2 kHz sonoreactor. Reprinted and adapted from [54], with permission from Elsevier.

tor. This is not only due to the nonlinear nature of the problem (further discussion on this topic is later found on this paper), but also due to the assumptions and limits of the models previously commented. A critical issue is the bubble size distribution, which must be known in order to estimate the effective sound speed of the bubbly liquid [57]. In this sense, the works previously commented [47, 48, 49, 53, 54, 56] used an arbitrary Gaussian distribution where bubbles much larger than those experimentally observed where considered. And even the Caflisch equations present their own limitations. In their model, Caflisch et al. [36] did not only assumed that the fluid velocity is small enough to actually neglect convention, but also considered very small volume fractions of bubbles, as the Foldy method they followed is not applicable due to the direct interaction of local pressure fields. This latter issue with the Caflisch model is actually reflected on its linearization by Commander and Prosperetti [43], who found that their model worked very well up to volume fractions of bubbles of 1-2%, and only in cases where bubble resonance plays a negligible role, not being that the case of typical cavitation phenomena experimentally observed in sonochemical reactors.

2.3. Vibration of the solid boundaries

All the previously commented studies treated the solid boundaries of the reactor as infinitely rigid/soft walls. Even though these rough boundary conditions may work fine with simple reactor configurations, it is clear that a more rigorous approach must be made accounting for the deformation of the sonoreactor walls due to their interaction with the acoustic pressure in the liquid, as infinitely rigid/soft walls inherently assume that no propagation of sound through the walls takes place. This assumption is indeed not true, as observed in several experimental studies where the working solution is not directly sonicated by the ultrasonic source (e.g. sonochemical systems where the reaction vessel is immersed in a ultrasonic bath [58, 59]).

FEM studies dealing with the simulation of the acoustic field in different media accounting for the vibrations of the vessel for different applications can be traced back to the 1980's, including the calculation of the acoustic pressure in ultrasonic tanks [60]. A good example of how the calculation of the acoustic field inside a sonoreactor can be coupled with the vibration of the reactor walls is the work of Liu et al. [61], who analyzed the presence of washing objects in an rectangular cleaning bath. In their study, they calculated the acoustic modes in the system, observing their dependence on the location of the washing objects, their mechanical restraint conditions and their physical properties. Also, they observed that resonance frequencies changed when assuming fixed or free mechanical boundary conditions for the objects in the bath, concluding that this was due to the oscillation of the washing objects. Another interesting result in their work was that the larger objects would act as if they were part of the tub, leading to a more complex ultrasonic field inside the cleaning bath and becoming more difficult to predict the structure of the acoustic field.

More recently, Yasui et al. [62] also coupled the linear acoustic field with the vibrations of the reactor walls in a sonochemical reactor working at 100 and 140 kHz with PAFEC-vibroacoustics software. Their work clearly demonstrated that the vibrations of the solid walls are a really important parameter in order to simulate the pressure field inside a sonochemical reactor. In fact, although they observed that thin glass walls (2 mm thickness) acted as nearly infinitely soft solids (free boundary) while thick glass walls were nearly rigid solids, there still were great differences in accounting for the vibrations of the reactor walls (Fig. 3A). In addition, they took into account the attenuation of the acoustic field by means of introducing an attenuation factor equivalent to the coefficient previously employed by Dähnke et al. [44, 47], Dahlem et al. [34] and many others. The inclusion of such parameter in the model allowed them to predict pressure antinodes where cavitation was experimentally observed, as shown in Fig. 3B.

In order to illustrate how the vibration of the solid walls of the reactor can be accounted for in the simulation of the acoustic pressure field, we will examine now the approach followed by Louisnard et al. [63] to couple the acoustics in the liquid and the deformation of the vessel in a deeper way. This

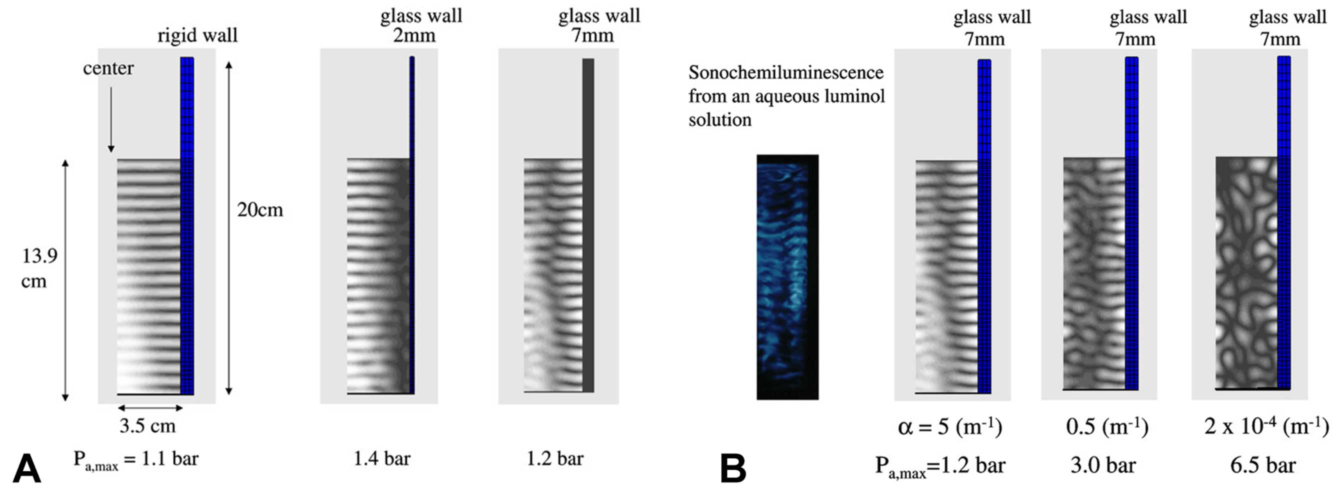

Figure 3: (A) Spatial distribution of the acoustic amplitude for a rigid wall and two glass walls (2 mm and 7 mm in thickness). The full-width at half maximum for the Gaussian distribution of the vibration amplitude of the vibrating plate is 5 cm. The attenuation coefficient is 5 m^{-1}. The wall height is 20 cm, while the liquid height is 13.9 cm. (B) Calculated spatial distribution of the acoustic pressure amplitude for a 7 mm thick glass wall for various attenuation coefficients in a 100 kHz reactor equipped with a vibrating plate transducer. The photograph of sonochemiluminescence from an aqueous luminol solution shows the corresponding half plane. Reprinted and adapted from [62], with permission from Elsevier.

linear-based model proposed a new approach to represent the ultrasonic horn in contact with the liquid in order to resolve the Helmholtz equation: assuming a horn radiating surface normal displacement of amplitude U_0, the inward acceleration of the liquid at the emitter surface of the horn can be defined as $\nabla^2 P \cdot \mathbf{n} = \rho_l \omega^2 U_0 \cdot \mathbf{n}$, being ρ_l the density of the irradiated liquid, while the lateral wall of the horn would be again considered as an infinitely rigid boundary. The vibration of the reactor walls were accounted for with the next expression, which was obtained by using the elasto-dynamic theory, neglecting volume forces and assuming mono-harmonic vibrations and the elastic deformation for all the materials employed in the construction of the reactor:

$$-\rho_s \omega^2 \mathbf{U}_s = \nabla \cdot \bar{\bar{\Sigma}} \tag{11}$$

where

$$\bar{\bar{\Sigma}} = \frac{E\nu}{(1-2\nu)(1+\nu)} \left(\mathrm{Tr}\, \bar{\bar{\xi}} \right) \bar{\bar{\mathbf{I}}} + \frac{E}{(1+\nu)} \bar{\bar{\xi}} \tag{12}$$

and

$$\bar{\bar{\xi}} = \frac{1}{2} \left[\bar{\bar{\nabla}}\, \mathbf{U}_s + {}^{\mathrm{T}}\bar{\bar{\nabla}} \mathbf{U}_s \right] \tag{13}$$

In eq. (10-12) , ρ_s is the density of the solid, $\mathbf{U}_s$ is the complex amplitude of the displacement field in the solid, $\bar{\bar{\Sigma}}$ is the elastic stress tensor, E is the Young modulus, ν is the Poisson ratio, Tr is the trace operator, $\bar{\bar{\xi}}$ is the strain tensor, and $\bar{\bar{\mathbf{I}}}$ is the identity tensor. In order to couple the Helmholtz equation with the vibration of the reactor walls, the next boundary conditions were defined by the authors:

- In the vessel walls where the liquid was in contact with a solid, the liquid acceleration must match the acceleration of the solid, and therefore $\nabla P \cdot \mathbf{n} = \rho_s \omega^2 \mathbf{U}_s \cdot \mathbf{n}$ was used as a boundary condition for the Helmholtz equation.
- To solve equation (2), a dynamic condition $\bar{\bar{\Sigma}} \cdot \mathbf{n} = -P \cdot \mathbf{n}$ was set where the solid was in contact with the liquid, stating that the normal force per unit area exerted on the solid boundary in contact with the liquid was just the pressure of the liquid, while the rest of the solid boundaries in contact with air were assumed free, and therefore the three components of the stress $\bar{\bar{\Sigma}} \cdot \mathbf{n}$ were ascribed to 0.

In their simulations, Louisnard et al. [63] considered a perfectly elastic reactor formed of external, non-dissipative and unconstrained elastic solid boundaries. In addition, they set an arbitrary, spatially uniform attenuation coefficient analogous to the coefficients previously used by other authors [34, 44, 47, 62]. This approach allowed the authors to obtain the response curves of the system as a function of frequency, pointing out the global resonance peaks of the whole mechanical system formed by the irradiated liquid and the walls of the reactor, showing how different was the response of the system when properly taking into account the deformation of the reactor walls (Fig. 4A). In this sense, the results clearly showed that interfaces between liquid and the solid walls cannot be properly represented by the simple approximations of infinitely soft/rigid boundaries, demonstrating that assumptions such as those made by Sáez et al. [18] (reactor walls defined as infinitely hard boundaries), or Klíma et al. [21] and Sutkar et al. [28, 29] (reactor walls defined as infinitely soft boundaries) and many others may yield inaccurate results for complex geometries. Liquid-solid coupling enabled the sound transmission to the cooling jacket (Fig. 4B), where the cooling liquid received part of the input power, opening the perspective to future designing of sonochemical reactors where the working liquid might not necessarily be in contact with the transducer emitter surface. This model has been further employed by Tudela et al. [64] to study the effect that the internal geometry may exert on the acoustic field in a sonoelectrochemical reactor, showing how the deformation of the electrode and the formation of alternated pressure antinodes on both surfaces of the electrode is perfectly coupled. The results of this recent work show how a proper election of both the geometry and the working frequency may enable to reach large acoustic pressure amplitudes, locating pressure antinodes far from the transducer and near the electrode. More recently, Harzali et al. [65] have used the model to obtain the response curves for a sonoreactor employed for the sonocrystallization of $ZnSO_4 \cdot 7H_2O$ at different solution heights, observing that the power dissipated and the induction time were well correlated as the liquid height was varied inside the sonoreactor.

3. Numerical simulations of the acoustic field inside sonochemical reactors using nonlinear-based models

Even though the acoustic pressure attenuation or damping effect (and implicit energy dissipation) due to the presence of cavitating bubbles has been accounted for in some of the previously mentioned studies, the different methodologies followed in all those works were basically linear-based approaches to actually solve a strongly nonlinear acoustic field where nonlinearity specially comes from the formation, growth and collapse of the cavitating bubbles. Nevertheless, as argued by Harvey and Gachagan [66], the nonlinearity appears when cavitation begins, which facilitates the use of linear acoustics-based models to predict pressure antinodes where cavitation would be likely to occur. The nonlinear propagation of sound is therefore inherent to acoustic cavitation, which in essence is a diphasic problem where bubbles rise from nowhere, arranging themselves in a wide variety of structures [67]. The complexity of the matter, related to the large range of spatial and temporal scales involved in ultrasonic cavitation (from the submicronic hot core in the bubble to the few centimeters wavelength; from the collapse of bubbles that may occur in a few picoseconds to the bubble structures observed during experiments that evolve and change their macroscopic features within few seconds), and the absence of a definite theory to explain most of the features of cavitation fields makes prediction and scaling-up of cavitation a hard task from an engineering point of view (for more information on the fundamental physics of cavitation, we strongly recommend further reading of Ref.[57, 68, 69]).

Despite the complexity of the problem and the lack of definite equations, a few groups have deeply investigated the matter in order to simulate a nonlinear acoustic field inside a sonoreactor. In a recent work, Louisnard [70] proposed a simple but powerful model which couples the evolution of both cavitation and acoustic fields by solving a nonlinear Helmholtz equation accounting for the bubble dynamics, the energy dissipation per bubble (where both thermal diffusion from the bubble towards the surrounding liquid and viscous friction in the liquid due

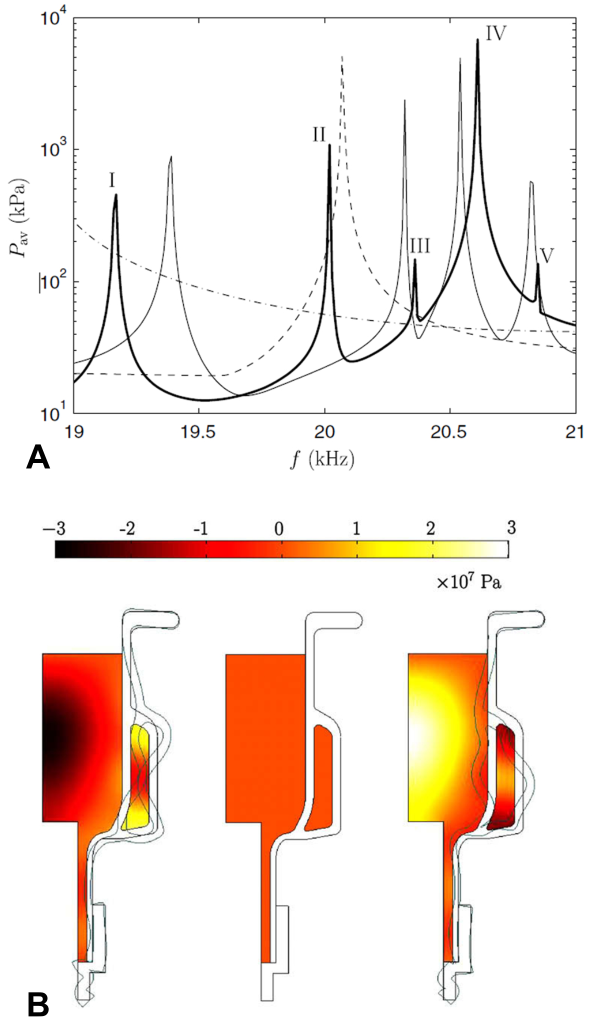

Figure 4: A: Response curves of the sono-reactor. Thin solid line: cooling jacket empty. Thick solid line: cooling jacket filled. Dashed line: solid boundaries considered infinitely rigid. Dash-dotted line: solid boundaries considered as infinitely soft. B: Pressure field $p(r, z, t)$ and wall deformation in a jacketed sonoreactor near a resonance frequency of 20610 Hz at times $\omega t = 0$, $\omega t = \pi/2$ and $\omega t = \pi$ when no attenuation is considered. The wall displacement is magnified 100 times. Reprinted from [63], with permission from Elsevier.

to its radial motion around the bubble are included), the energy conservation in the liquid and the nonlinear attenuation of acoustic waves. Although this approach does not solve the fully nonlinear Caflisch equations (which would require extremely complex temporal integration to reach a solution [71]) and its based on the consideration of unproven approximations in the derivation of the nonlinear Helmholtz equations, it provides quite more realistic acoustic pressure amplitudes than those previously obtained by fully linearized models [63]. Moreover, this model predicts a strong attenuation due to the presence of the bubbles which produces a traveling wave component even in a closed system with perfectly reflecting boundaries (a feature that was already observed with the previous linear-based approach [64] when using an attenuation coefficient) and that could explain the traveling wave and the standing wave fields observed by Son et al [72] in their cylindrical reactors. This attenuation has a strong influence on the magnitude and orientation of the primary Bjerknes force and on the bubble structures [73], where the introduction of the nonlinear Helmholtz equation in the previously developed model accounting for the vibration of the boundaries [63] enables to predict commonly observed cavitation phenomena such as conical bubble structures under ultrasonic horns [74] (Fig.5) or flare structures very similar to those experimentally observed in ultrasonic cleaning baths [67]. This actually constitutes a strong feature of Louisnard's model, as linear-based models completely fail to predict the location of pressure antinodes in cases as the cone bubble structure widely observed in the emitter surface of horn transducers. However, the model presents two main drawbacks, which are the arbitrary choice of the ambient radius of the bubbles and the homogeneous bubble density distribution in the regions where the pressure is above the Blake threshold, an issue that may be enhanced in a short term basis by properly treating the spatial redistribution of the cavitating bubbles by means of coupling the nonlinear Helmholtz equation with a convection-like equation for the bubble number density. Still, Louisnard's simulations successfully demonstrate how the spatial distribution of the acoustic pressure in cavitational fields is deeply controlled by strong energy dissipation due to the bubbles.

Vanhille and Campos-Pozuelo (and co-workers) have also deeply investigated the theoretical and mathematical development of different models to calculate the propagation on nonlinear ultrasonic waves in fluids. From previous models formerly developed for (i) the analysis of the mean acoustic pressure coupling the fluid mechanics and Rayleigh-Plesset equations [75] and for (ii) the propagation of ultrasound coupling the acoustic field and the vibrations of bubbles [76], they numerically investigated the time-dependent propagation of nonlinear ultrasonic waves in bubbly liquids with a non-homogeneous distribution of bubbles [77] and the physical mechanism at the origin of the commonly observed conical bubble structure under ultrasonic horns [78] in the 1D semi-infinite space domain, where bubbles were concentrated in some regions. This model, based on former theoretical postulations by Zabolotskaya and Soluyan [79], considers that both the nonlinear behaviour and the attenuation of the acoustic field are only attributable to the nonlinear oscillations of the bubbles. This nonlinear nature of the bubbles comes from the nonlinearity shown by the state equation of the gas in the bubble, which is defined as adiabatic, and from the spherical symmetry of the problem. By defining the gas in the bubble as adiabatic, the authors assume that there is no heat transfer between the bubbles and the surrounding medium. This assumption, although it could be used safely in some cases, may sound controversial when considering the attenuation of the acoustic field inside a sonochemical reactor, as heat diffusion from the bubble towards the liquid does occur, which results in the attenuation of the acoustic wave to some extent (a rough estimation for the heat diffusion from the bubble towards the liquid was proposed by Prosperetti et al. considering the continuity of the heat flux at the bubble [80], more recent treatments of this issue can be found in works by Storey and Szeri [81, 82] and Toegel et al. [83]; previous work from Louisnard [70] showed that, although wave attenuation is mainly governed by viscous dissipation coming from the radial motion of the bubble, thermal effects also play a minor role). Another drawback in their model is viscous damping, which is linear-based and results in minor wave attenuation, as opposed to the observations made by Louisnard [70]. Despite these considerations, their model successfully showed that the nonlinear nature of the bubbly liquid excited by a 24.5 kHz transducer in a rectangular cavity yielded the generation of higher harmonics at high amplitudes taking into account the dissipation and the dispersion due to the presence of the bubbles [84]. Their model is therefore able to simulate the nonlinear behavior of ultrasonic waves in reactors where a bubbly liquid is placed. More recently, they have introduced new features in their model related to the bubble formation process [85]. In this mainly theoretical latter work (all the calculations are in one-dimensional domain), although they neglected physical phenomena such as

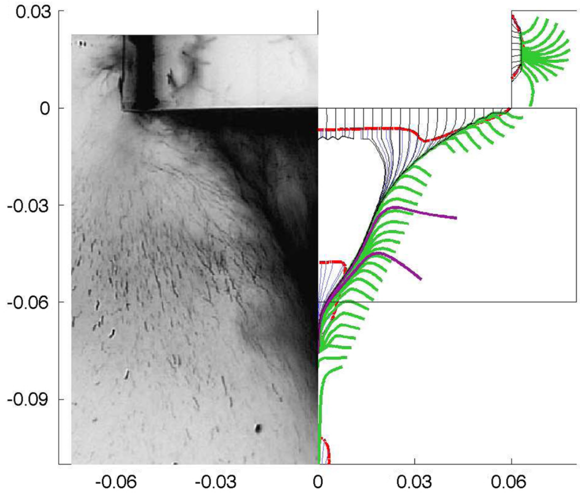

Figure 5: Comparison between (right) numerical simulations [73] and (left) experimental results [74] of a conical bubble structure under an ultrasonic horn, where the black lines are the S-streamers, the blue lines are the L-streamers, the thick solid red line is the Blake threshold contour curve, and the green and magenta lines show additional streamers originating from arbitrary points points below the Blake threshold (for interpretation of the references in colour in this figure legend, the reader is referred to the web version of this article). Reprinted and adapted from [73] and [74], with permission from Elsevier.

primary and secondary Bjerknes forces and acoustic streaming, and they defined small bubbles which acted as nuclei in the initial state of the fluid (implying that the cavitation threshold is lower than the Blake threshold), they demonstrate how their updated model not only provides an equation that successfully shows a mechanism of bubble generation, but also how the bubbles generate the nonlinearity that affects the acoustic field inside a sonochemical reactor. It must be noted though that some of the assumptions made by Vanhille and Campos-Pozuelo in the latter work are unrealistic compared to real experiments where ultrasonic cavitation occurs: the maximal volume fraction of bubbles was set at 0.0038%, while the cavitation threshold was set at 8000 Pa, which is indeed very small compared to real cavitation phenomena.

4. Simulation of the ultrasonic transducer

In the previous section, accounting for the vibration of the solid boundaries was briefly commented, concluding that the deformation of the reactor walls effectively affects the acoustic field and, therefore, it should not be neglected. In this sense, a question rapidly comes up: why not considering the vibration of the ultrasonic transducer too? Most of the works found in the literature usually treat the emitter boundary as a acoustic field source where a uniformly distributed acoustic pressure, velocity or displacement (usually introduced as a normal acceleration condition) is assumed, ignoring the effect that the deformation of the emitter surface may exert on the acoustic field. As an example, the modal vibration of transducers could not only affect the acoustic field near the emitter and therefore the streamer patterns at its surface (as those observed by Moussatov et al. [74] for a 20.7 kHz sonotrode), but also the lateral cavitation fields commonly observed when a low frequency ultrasonic horn is submerged in the working liquid (as the streamers in the lateral wall of the ultrasonic horn formed by its radial motion observed in the simulations by Louisnard [73], Fig. 5). Horst et al. [86] were among the first researchers who theoretically investigated the fundamentals on the energy conversion paths in a sonochemical reactor designed for heterogeneous reactions where coupling between the irradiated liquid and the transducer was evaluated by a 4-pole analysis of stepped transducer horns assuming a basic linear vibration of the bubbles. Since then, only a few works have actually tried to couple the vibration of the ultrasonic transducers and the acoustic field inside a sonoreactor. Among these, we may find a quite interesting work by Cancelos et al. [87] where they defined a basic mathematical model with the ATILA code to design acoustic resonant chambers. Perincek et al. [32] also seemed to account for the vibrations of the emitters, although no further details about their approach to the problem were included in their paper. And Gachagan et al. [94] simulated a rectangular cell equipped with a 40 kHz transducer with 2D FEM analysis with the Ansys software package (again, no detailed information about the specific model they used is given in the paper). In their work, they were able to predict cavitation areas on different horizontal levels around the center of the cell which were roughly in agreement with their experimental work.

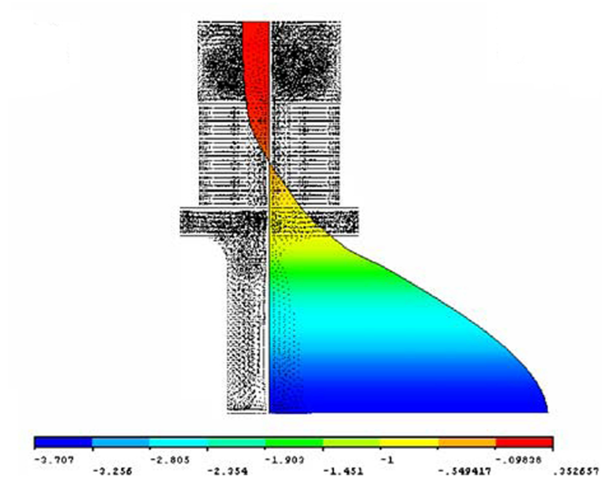

Figure 6: Location of node from modal analysis in full 3D mesh of a 17.2 kHz ultrasonic transducer. Reprinted and adapted from [90], with permission from Springer.

A straightforward reason for the lack of theoretical works on the interaction between the ultrasonic transducer and the irradiated fluid in the literature is the lack of information on the internal design of the ultrasonic transducer as no information is usually given by the suppliers on the specific working parameters of the piezoelectric ceramics, back mass and amplifier of the commercial ultrasonic transducers. In fact, diverse research groups have been dealing with similar issues for a quite long time, and some works focused on the modeling and designing of ultrasonic transducers with different purposes are found in the literature [88, 89, 90, 91] (Fig.6). Quite recently, Memoli et al. coupled the vibrations of the transducer and the pressure field in the irradiated liquid [92, 93]. In their works, the authors used the Ansys FEM code to study the effects of temperature changes on the operation of a 25 kHz sonoreactor by using a linear acoustics-based model previously developed by Birkin et al. [97]. The transducers in these studies were modelled using piezoelectric orthotropic elements where the all the piezo-elastic properties were considered (elastic, piezoelectric and dielectric constants). To implement the piezoelectric effect in the simulations, we need to relate the stress and strain with the electric field and electric displacement. For this purpose, we can either used stress-charge or strain-charge equations. The equations in the stress-charge form are:

$$\bar{\bar{\Sigma}} = \mathbf{c}_E \, \bar{\bar{\xi}} - \mathbf{e}^{\,T}\mathbf{E} \tag{14}$$

$$\mathbf{D} = \mathbf{e} \, \bar{\bar{\Sigma}} + \epsilon_0 \, \epsilon_{rS} \, \mathbf{E} \tag{15}$$

where $\mathbf{E}$ is the electric field, $\mathbf{D}$ is the electric displacement, ϵ_0 is the electrical permittivity of free space and $\mathbf{c}_E$, $\mathbf{e}$ and ϵ_{rS} are the piezoelectric elasticity matrix of the ceramic, the coupling matrix and the relative permittivity for the stress-charge equations, respectively. The equations in the strain-charge form are:

$$\bar{\bar{\xi}} = \mathbf{s}_E \, \bar{\bar{\Sigma}} + \mathbf{d}^{\,T}\mathbf{E} \tag{16}$$

$$\mathbf{D} = \mathbf{d}\,\bar{\bar{\xi}} + \epsilon_0\,\epsilon_{rT}\,\mathbf{E} \tag{17}$$

where $\mathbf{E}$, $\mathbf{d}$ and ϵ_{rT} are the piezoelectric elasticity matrix of the ceramic, the coupling matrix and the relative permittivity for the strain-charge equations, respectively. As the electric field is defined by $\mathbf{E} = -\nabla E$, being E the electric potential, it is possible to account for the whole electromechanic chain by just setting the applied voltage between the piezoceramic terminals, as shown in Fig. 7 for some initial tests carried out by the authors of the present review with COMSOL Multiphysics software using the linear-based model previously described [63].

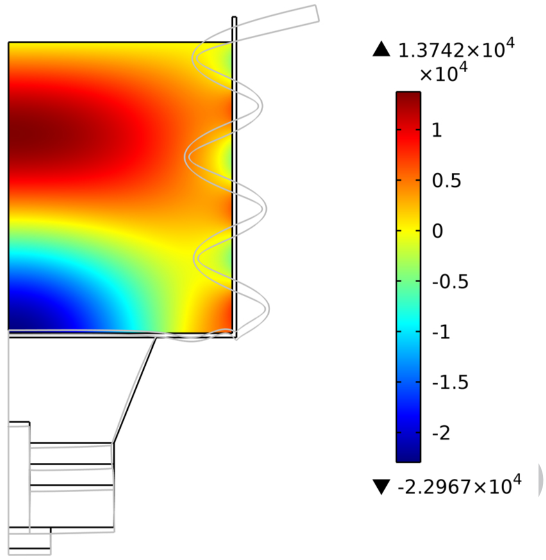

Figure 7: Pressure field $p(r, z, t)$ (in Pa) and solid vibration in a axisymmetric ultrasonic bath equipped with an ultrasonic transducer equipped with two piezoceramics near a resonance frequency of 19240 Hz at $\omega t = 0$ when no attenuation is considered. The wall displacement is magnified 100000 times.

5. New challenges and trends in coming years

Even if the simulation of the whole electromechanical transducer can be accounted for in a relatively easy way, we have to keep in mind the complexity of rigorously modeling the electromechanic transducer, the vibrations of the reactor walls, and the acoustic field inside the reactor. Regarding the electromechanic transducer, pre-stress must be accounted for in order to avoid the estimation of resonant frequencies that may significantly differ form the real one. It must be also taken into account that nonlinearities can occur in the solid deformation in both the transducer and the reactor, resulting in different nonlinear effects such as frequency shifts, hysteresis phenomena, amplitude saturations and modal interactions [95, 96]. Development of a more rigorous acoustics model accounting for the nonlinear propagation of sound waves and the attenuation of the sound pressure by cavitation is also necessary. Related to this, the apparition of new nonlinear models as those previously mentioned seems quite promising for new simulation strategies and the development of new designing procedures from an engineering point of view, as opposed to the use of linear-based models that do not properly account for the attenuation coefficient in a cavitating liquid, even when implementing the linearization of the Caflisch equations [43] into the Helmholtz equation. This results in linear-based models failing at showing the self-attenuation of the acoustic field that takes place in real sonochemical systems, as demonstrated by Campos-Pozuelo et al. [75] and theoretically discussed by Louisnard [70], and is linked to the assumption of linear oscillations for cavitating bubbles, which is indeed far from reality. Therefore, although some simulations carried out with linear-based models may show certain agreement with some experimental results, these are more qualitatively than quantitatively-based, and in all cases, based on simple geometries and reactor configurations, so it is important that researchers willing to start working on the field are aware of the current limitations of the numerical simulations of the acoustic field inside any sonochemical reactor.

Besides this, there is a straightforward question related to the numerical simulation of the cavitation field inside a sonoreactor: What about the bubbles themselves? How could we calculate a diphasic cavitation field where the spatial bubble distribution forms complex structures which actually vary on a timescale much longer than the acoustic period? In a recent discussion on this topic, Louisnard and González-García [57] briefly reviewed the continuum and the particle modeling approaches. The first approach, which assumes bubble distribution in a sonoreactor as a continuous function of space, time and bubble size (its evolution would be described by a bubble conservation equation), is mainly theoretical and outstandingly complex, although different sets of equations have been proposed by different authors [98, 99] (for those readers willing to know more about this complex topic, we strongly recommend to check the detailed discussion made by Louisnard [71]). And the second approach, which would treat individual bubbles as particles under the influence of different forces (primary and secondary Bjerknes forces and added mass and viscous drag forces [57]), presents quite more interest from an applied point of view, as it could easily explain the formation of bubble structures and streamers. Regarding the use and development of particle models in order to qualitatively simulate the different bubble structures commonly observed in ultrasonic cavitation phenomena, we strongly recommend the reader to check some of the works by Mettin and co-workers [55, 67, 100], where they developed different particle models which enable to simulate bubble structures close to those observed in real experiments as shown in Fig 8. Coupling these particle models with nonlinear models as those commented in this review could therefore lead to a more rigorous simulation of the acoustic field and the different time-dependent bubble structures commonly observed in sonochemical reactors, and the heterogeneous nature of any sonochemical system (liquid and bubbles) would be even better reflected in the simulation.

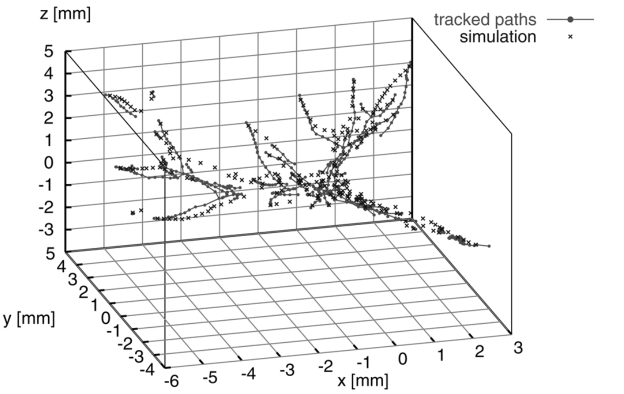

Figure 8: Three-dimensional comparison of experimentally recorded (lines with dots) and simulated (crosses) bubble tracks. The temporal distance between successive dots (or crosses) of a track corresponds to 440 μs. Reprinted and adapted from [100], with permission from Elsevier.

6. Acknowledgment

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Simulation of the spatial distribution of the acoustic pressure in sonochemical reactors with numerical methods: A review

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Highlights:

- This paper summarizes some of the work developed in the last 15 years on the simulation of the acoustic field inside sonoreactors by numerical methods.
- Recent works on this topic, including those focused on the vibration of the reactor walls and the nonlinear phenomena inherent to the presence of ultrasonic cavitation, are briefly illustrated and discussed.
- This review also comments some of the upcoming challenges that must be addressed in order to develop numerical simulations of the acoustic field inside a reactor as a valuable tool for an efficient design of sonoreactors.